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Final Progress Report

1. Foreword

This MURI grant has funded the pursuit of a fundamental research effort designed to aid United States combat forces in reducing their visibility to opposing forces over a broad electromagnetic spectrum extending from the millimeter wave range to the visible and near infrared. This work sought to enable the fulfillment of the Full Dimensional Protection strategy by providing new, robust technological solutions for achieving limited visibility operations across a significant portion of the electromagnetic spectrum.

Metal Nanoshells, novel nanoparticles fabricated with ultrathin conductive shell layers, provide the scientific breakthrough at the heart of this proposal. By varying the relative thicknesses of the core and shell layers, the electromagnetic response of the nanoparticles can be sensitively tailored across the visible and infrared regions of the spectrum. Simple variations of this basic core-shell structure extend the controlled electromagnetic response of these nanoparticles into the far-infrared and millimeter-wave regions. Chemical functionalization of these nanoparticle surfaces permits their attachment to a wide variety of surfaces by simple and robust chemical methods. The addition of a voltage-tunable guided-wave structure beneath the nanoparticle surface layer or layers renders the nanoparticles adaptive in nature, that is, capable of a voltage-dependent optical response. This simple planar geometry permits the detailed study and optimization of this device; it also permits scalability to cost-effective, large surface area structures and high volume production.

Metal Nanoshells possess several distinct advantages over photonic bandgap structures- there is no need to fabricate large, defect-free periodic structures to achieve the predicted optical properties- the optical properties are designed into the individual nanoparticles themselves. This is an overwhelming advantage, because it permits the straightforward development of both passive coatings and active devices with low observability signatures in multiple regions of the electromagnetic spectrum simultaneously, for example, the infrared and millimeter-wave regions. The nanoscale dimensions and easy chemical functionalization of metal nanoshell surfaces permits their incorporation into a wide variety of media: plastics, epoxies, liquids, aerosols, glasses, even fabrics. Metal nanoshells are remarkably strong optical scatterers as well as absorbers, and can be designed to scatter light *with no backward scattering* over specified frequency ranges. Metal nanoshells also demonstrate a highly sensitive chemical-specific optical response due to the enormous Raman Scattering enhancements available at their surfaces. This property can be advantageously exploited in the development of new, all-optical *in vivo* biosensing and bioassaying strategies that can be tailored for military or commercial applications.

To fulfill the goals of this grant we pursued a comprehensive research program that advanced the development of nanoshells and closely related nanoparticle structures for use as both passive and adaptive optical materials with highly controllable electromagnetic responses from the visible to the millimeter wave region of the spectrum. The effort is both multidisciplinary and multi-institutional, involving researchers at Rice University (Departments of Physics, Electrical and Computer Engineering, and Bioengineering), the University of Houston (Department of Chemistry), and Oklahoma State University (Department of Electrical and Computer Engineering). The proposed research program was based on several successful current collaborations among the members of the research team previously and concurrently funded by the Department of Defense, the National Science Foundation, and the National Aeronautics and Space Administration. Our research program constitutes a strong and coordinated integration of nanoparticle design and synthesis, film and device fabrication, characterization of optical and electromagnetic properties over the infrared and terahertz spectral regions, theoretical analysis, and commercializable applications.

2. Table of Contents (if report is more than 10 pages)

3. List of Appendixes, Illustrations and Tables (if applicable)

4. Statement of the problem studied

The specifically stated goals of this project were:

- The design and fabrication of nanoshell particles and closely related nanoparticle assemblies with a tailored and optimized optical/electromagnetic response in the near-, mid-, far-infrared and submillimeter-wave spectral regions;

- The development of films and thin-film device structures with passive and/or active control of electromagnetic properties in any region or regions of choice from the visible to the millimeter-wave;

- The characterization of the nanoshells and nanoparticles fabricated in this research, and the proposed films and devices, using a broad range of experimental techniques particularly suited to the study of their optical, electronic, electromagnetic, and photophysical properties. These methods include:

  (a) relevant optical spectroscopies, such as UV-vis, FTIR, FT-Raman, photoluminescence, light scattering and nonlinear optical characterization *via* degenerate four-wave mixing, transient absorption, and optical Kerr effect measurements,

  (b) terahertz time domain spectroscopy and ranging, to probe passive, optical, and voltage-dependent submillimeter-wave response of nanoshell-constituent structures,

  (c) local probes for structural information on fabricated arrays, devices, and the constituent nanoparticles, such as atomic force microscopy, transmission electron microscopy, and X-Ray diffraction and scanning tunneling microscopy;

- Theoretical investigations and interpretations of individual nanoshell properties, such as optical excitation and applied field response, electromagnetic wave scattering properties, collective effects and interparticle interactions in nanoshell arrays, films, and device structures;

- The development of a nanoshell-based all-optical biosensor and bioassay based on the surface enhanced Raman response of molecules attached to the nanoparticle surfaces.

We originally anticipated that this research grant would allow us to develop a breakthrough technology that would enable the control and manipulation of infrared to submillimeter-wave electromagnetic radiation with unprecedented precision, based on components that can be ultimately produced *en masse* and at moderate cost. ***We can state that we have achieved success in accomplishing these goals.***

5. Summary of the most important results

This grant supported the experimental development of metal-coated silica nanoparticles known as nanoshells. The nanoparticle geometry consists of a dielectric core surrounded by a thin metallic shell of uniform thickness. In this geometry, the optical resonance, derived from the plasmon resonance of the metal which composes the film material, can be shifted in wavelength in a highly controlled manner. By varying the relative size of the dielectric core and the metallic shell the optical absorption resonance can be shifted or tuned to any selected wavelength ranging from the visible region of the spectrum to the far infrared, providing a unique tunability of this nanostructure over an extremely broad region of the electromagnetic spectrum.

Our major accomplishments in fabrication of this nanostructure involved the development of a reliable, solution based fabrication method that permits the growth of a gold layer of controlled thickness onto silica nanoparticles. This fabrication method was developed and extended to nanoparticles with resonances ranging from the visible region of the spectrum to the mid infrared range of the spectrum (2.9 microns). Silver nanoshell fabrication chemistry was also developed based on the same seeded growth method, but with less controlled growth than the gold electroless plating process due to the much more rapid kinetics of the silver electroless plating process. Encapsulating a gold nanoshell within a second silica layer and outer gold shell, a structure we called “nano-matryushka”, provided a particle with resonances that extended from the near infrared to the far infrared region of the spectrum (nominally 9 microns). The electroless plating method was also adapted to planar geometries using a combination of microcontact printing and electroless plating, to create patterned metallized surfaces that can be produced over large planar areas with submicron resolution by a simple mask-free stamping and developing process.

One of the major contributions of this MURI effort was the development of a theory for the plasmon response in finite metallic nanostructures of complex geometries such as the nanoshell. The plasmons supported by this geometry arise by a hybridization of the plasmon states of simpler fixed frequency structures: in this case the plasmon of a solid sphere and that of a solid cavity. The mixing of these two primitive plasmon states results in the formation of two nanoshell plasmon resonances, a low energy “bonding” plasmon and a higher energy “anti-bonding” plasmon, in direct analogy with molecular orbital theory. The lower energy “bonding” plasmon couples directly to light, and it is this plasmon that is used in virtually all experimental studies and applications. The plasmon hybridization picture is a general and far-reaching principle (because it is rigorously quantum mechanical) and applies in general to virtually all plasmonic geometries. We used the plasmon hybridization formalism to explain the nano-matryushka plasmon resonances as hybridized plasmon states form by mixing of the inner and outer nanoshell plasmons. This extremely powerful method is currently being used extensively by our group to determine the energy spectrum of a large variety of plasmonic geometries.

Physical properties of nanoshells were studied by numerous methods. Optical properties of nanoshells were studied both in the spectral domain and for their dynamical, time-dependent response. It was observed that the surface chemistry could modify the plasmon lifetime systematically, by the induced dipole moment of the adsorbate molecules. Although the plasmon can be tuned in this nanostructure, the nonlinear optical response was found to be controlled by the bulk density of states of the metal, which determine at which wavelengths one can observe photoinduced absorption vs. photobleaching. Studies of the nonlinear optical properties of nanoshells were found to be dominated primarily by photothermal effects. Under high intensity laser irradiation, the photoinduced destruction of the nanoparticle was examined, where nanosecond laser excitation resulted in a very different morphology than femtosecond excitation. Under nanosecond excitation the metallic shell layer reshaped to form small islands on the nanoparticle surface, a transformation accompanied by a dramatic color change due to the

reshaping of the gold into spherical structures with higher energy plasmon resonances. Under femtosecond laser excitation the gold shell layer was removed in a highly asymmetrical fashion, with the majority of the metallic shell being deposited on one side of the silica nanoparticle. These observations may prove relevant for fabrication of unusually shaped reduced symmetry nanoparticles. The thermal stability of nanoshells was studied, and it was observed that the shell layer melted at nominally 300 degrees Celsius, far below the bulk melting temperature of gold. Growth of a silica stabilization layer around the nanoshells elevated the melting temperature by more than 200 degrees. The growth of molecular layers on the surfaces of nanoshells was performed for the first time. Alkanethiol self-assembled monolayers (SAMs) were grown on gold nanoshells and dense films of the functionalized nanoshells were subsequently deposited on dielectric substrates.

The very strong photothermal response of nanoshells was exploited in the development of a new and novel optomechanical material. Nanoshells were dispersed into the photothermally responsive polymer co-N-isopropylacrylamide-acrylamide (NIPAAm-co-AAm). When this copolymer is heated above its lower critical solution temperature it undergoes a quasi-first order phase transition and a dramatic decrease in volume that is reversible upon cooling. The addition of nanoparticles to this material does not significantly change its thermal response, rather, it enables the triggering of this response by laser light when the laser wavelength corresponds to the resonant wavelength of the embedded nanoshells. This results in an optomechanical material that can be useful for photo-assisted drug delivery and for phototriggered microfluidic valves. We also demonstrated that plasmon resonant nanoparticles of differing wavelengths could be used to make this photoinduced collapse a wavelength specific response.

Nanoshell dopants also proved to be useful in enhancing and modifying the properties of other polymer materials. In conducting polymers, nanoshells were tuned to be resonant with the triplet exciton of the polymer, and proved to be an effective triplet state quencher in both MEH-PPV and P3OT. Quenching of the triplet state is extremely important in arresting the photodegradation of conducting polymers, and it was shown that extremely small quantities (0.1% by volume) of nanoshells could slow the oxidation of these polymers by a factor of ten in material lifetime without affecting the material's photoluminescence efficiency.

Since the optical absorption, or far field, optical response of nanoshells is controlled by nanoparticle geometry, the geometry must also control the near field optical response. Therefore the nanoparticle geometry controls the intensity of the field at the nanoparticle's surface. This field is responsible for the major enhancement mechanism of surface enhanced Raman scattering (SERS). We examined how we can tune and optimize the SERS response of nanoshells. In particular, it was found that nanoshells can be used to enhance SERS at near infrared wavelengths. In a set of initial studies we showed that the SERS enhancement for nanoshells was maximal just before the shell layer was complete, suggesting that local nanoscale roughness may provide part of the enhancement needed for SERS on nanoshell substrates. We examined both experimentally and theoretically how the core and shell dimensions of nanoshells could be optimized for the wavelengths 1.06 micron and 780 nm, and showed that the actual SERS response for nonresonant molecules agreed very well with electromagnetic theory for both of these wavelengths. In collaboration with Thomas Huser's research group at Lawrence Livermore National Laboratory, we performed a quantitative study comparing SERS on individual solid Au nanospheres, nanosphere dimers, nanoshells and nanoshell dimers all coated with a monolayer of nonresonant but SERS active molecules (paramercaptoaniline). In this study we showed that the SERS signal from a monolayer of molecules on an individual nanoshell was approximately as strong as that from molecules in a single nanosphere junction or "hot spot". This was the first

quantitative study on SERS substrates connecting theoretically calculated electromagnetic responses with the actual measured SERS intensities.

A nanoshell based immunoassay was developed that can be utilized to measure physiologically relevant quantities of analyte within whole blood, in just a few minutes, with no sample preparation. Nanoshells with plasmon resonances in the near infrared were conjugated with antibodies and dispersed in whole blood. In the presence of an analyte the nanoshells began to aggregate, giving rise to a strong redshift in their plasmon resonance due to the aggregate plasmon. This change in optical properties could be monitored using near infrared light at wavelengths where blood is transparent.

A novel "texturing" chemistry for nanoshells was discovered, and used to prepare nanoshells with intentionally roughened surfaces. A simple chemical, cysteamine, that slowly etches solid Au nanocrystals, was found to preferentially etch nanoshells at the domain boundaries between the crystal grains on their surfaces. The plasmon resonant response of these structures were studied both experimentally and theoretically. Remarkably, it was observed experimentally and verified theoretically that the plasmon resonant response is very robust with respect to surface roughness. The addition of surface roughness will damp out the higher order plasmon modes (such as the quadrupole or octupole modes when present due to particle size) but the nanoshell must be severely disrupted (i.e. deep fissures in the shell layer, or other major discontinuities) for the dipole plasmon to be affected. On a continuous but rough nanoshell the addition of roughness adds approximately a factor of 2 to 5 in field enhancement relative to a perfectly smooth nanoshell; this provides an increase in SERS enhancement of approximately 1 to 2 orders of magnitude.

A detailed study of the relative contributions to the plasmon linewidth of metallic nanoshells was performed. In addition to inhomogeneous broadening effects due to size variations in both core and shell dimensions, other effects were investigated such as phase retardation (increase in particle size with respect to the incident wavelength of light) and also "electron scattering", a controversial mechanism long thought to be responsible for the broad linewidth of the plasmon lineshape. In a detailed study it was found that the predominant effect in determining lineshape for nanoshells was phase retardation. Following our initial study we performed a single particle spectroscopic study of nanoshells and determined that the lineshape of individual nanoshells fit Mie scattering theory exactly, without the need to invoke an electron scattering mechanism to explain the linewidth. Further study beyond these two investigations is warranted to explain the difference between the single particle plasmon lineshape and that observed in ensemble measurements.

We also reported the fabrication and optical properties of reduced symmetry nanoshells, "nanocups" and "nanocaps". These were fabricated by depositing nanoshells onto a surface electrostatically, performing nanoshell metallization chemistry, and then removing the asymmetric nanoparticles using an ultrasonic probe. The electromagnetic properties of these structures were determined and the optical response of the oriented nanoparticles was studied both experimentally and theoretically with good agreement. In a related experiment, it was discovered that exposure of nanoshells to the surfactant CTAB (cetylammonium bromide) could slowly reshape and remove the gold shell layer, morphing the nanostructure into unusual and highly regular shapes that cannot be fabricated by other methods. The CTAB is also etches the silica core slowly over time. The structures that result are (1) cylindrical, beanlike structures, and (2) highly regular, "bagel"-like nanostructures.

We performed a comprehensive study of the sensitivity of the surface plasmon resonance (SPR) wavelength of nanoshells to changes in the dielectric function of their embedding medium (refractive index of solvent) both theoretically and experimentally. While the sensitivity of SPR shift to refractive index increases with total size of the nanoparticle it is not particularly sensitive to core-shell ratio. We measured the response of nanoshells as SPR sensors across a wide range of refractive index changes. These measurements were performed by dispersing nanoshells onto a dielectric support using polyvinylpyridine, a highly successful attachment strategy which produces films of nanoshells with virtually no aggregation, preserving the outstanding optical properties of nanoshells in solution. These nanoshell films were quite robust to solvent changes and provide a practical strategy for making nanoshell SPR sensors that could be used for a wide variety of applications and that are cheaply and straightforwardly manufacturable.

We also investigated nanoshells and other nanopatterned metallized structures in planar geometries. We studied the light coupling of nanoshells with a planar metal substrate, and the variation of this coupling with distance between the nanoshell layer and the metallic plane. We also studied the propagation characteristics of surface plasmons on metallodielectric grating structures fabricated by the microcontact printing-metallization method we developed. The plasmonic band gap of these structures was sensitive to changes in dielectric environment and in chemical functionalization, and these studies were reported.

Experimental efforts have focused on THz Time Domain Spectroscopy (THz-TDS) of systems where a small change in some externally controllable parameter can potentially yield a large change in far infrared optical properties, THz imaging systems, and development of spectroscopic measurement systems for thin films.

These efforts include fundamental science of pulse propagation near the critical angle in dielectrics, and THz characterization of polymers and nanoshells. These systems are of interest to the overall goals of this project as potential systems for metal insulator transitions in the THz spectral region. Additionally the optical properties of nanoshells are dependent upon the bulk dielectric properties of the shell material, which can be sensitively measured with THz-TDS.

We have made time-domain measurements with subpicosecond resolution of optical tunneling of THz electromagnetic pulses undergoing frustrated total internal reflection. The transmission across a broad bandwidth is shown to be sensitive to small changes in incidence angle and optical barrier thickness. Measurements were additionally done over a range of incidence angles, through a new spectroscopic technique developed during this project.

A variety of material systems have been investigated. Since electrical characterization of conducting polymers requires fabricating ohmic contacts we have applied THz-TDS to directly measure both the absorption and dispersion of conducting polymer films. These include various polymer systems: polypyrrole with very high carrier concentrations, and polyaniline with much lower carrier concentrations. We have also investigated protonated polyaniline systems grown at University of Houston and films of low conductivity poly-3-methylthiophene, prepared electrochemically at -40^{o} C from low frequencies to beyond 4 THz. The complex conductivity, determined over the full frequency range, was 30 times lower than previous THz-TDS measurements of the 215/(Ω cm) high conductivity polypyrrole. Polypyrrole was well fit by Drude theory, however for poly-3-methylthiophene the measurements are fit by a localization-modified Drude theory.

Preliminary investigations of thin films of metal nanoshells on silicon substrates showed a strongly frequency dependent structure in the THz region. While the real and imaginary parts of

the refractive index show behavior qualitatively similar to conducting polymers as shown below, the measured index does not fit the Drude theory. Use of the localization modified Drude theory provides better fits, however much behavior is still unexplained, including the sharp index feature at 250 GHz.

To clarify the conduction processes in these materials as a function of external perturbation, we have focused on developing new measurement techniques. This project developed a THz interferometer which is extremely sensitive to small changes in sample composition and additionally removes unwanted experimental artifacts particularly in the case the sample is very thin, or with low index and absorption when it is very difficult to distinguish changes in THz pulse caused by the sample from those caused by long term fluctuations in the driving laser source or experiment. This instrument, the only one of its type in the world, has been used for characterization of numerous thin film materials. This system automatically compensates for changes in the THz signal over the course of a measurement due to slow laser fluctuations allowing near real time measurements of the film thickness-index product with a resolution of under one micron.
For typical values of thin films (d 1 m, n_r 2, 1%) the increase in measurement accuracy over THz-TDS is on the order of a factor of 50. With the minimum detectable signal on the order of 0.2 pA, this corresponds to a time resolution of 1 fs or the ability to detect films of thickness on the order of 100 nm. We are currently working to develop THz systems with higher bandwidth and correspondingly better sensitivity.

A key problem in THz measurements, especially phase coherent imaging measurements is knowledge to the spatio-temporal distribution of electric field phase. To determine the spatio-temporal field distribution of freely propagating THz bandwidth pulses we have investigated the time resolved electric field in two spatial dimensions with high spatial and temporal resolution. We have compared field measurements favorably with theory using both a simple plane wave and FDTD methods. The effects of both spherical aberration as well as surface wave coupling play important roles in phase front propagation, and are observed directly in the time domain. Measurements of the complex spatial amplitude distribution of optoelectronically generated THz beams indicate that the beam does not behave like a TEM_{00} Gaussian. THz beams can exhibit complex spatial profiles, particularly in the near field or in image planes, which must be considered in applications of lens coupled THz sources where field patterns are important, such as ranging or coupling into waveguides.

This grant supported the development of theoretical methods for description of the electronic and optical properties of metallic nanostructures. Our major accomplishments include the development of an ab initio TDLDA method for the quantum mechanical description of optical response of spherical nanoparticles, the development of an efficient method, “The plasmon hybridization method” for the direct calculation of plasmon energies of complex metallic nanoparticle structures, and the development of an efficient parallel Finite-Difference Time-Domain (FDTD) method for simulation of the electromagnetic properties of arbitrary nanostructures. The methods developed have been used both to stimulate experiments as well as analyze experimental data in several joint theoretical-experimental studies.

6. Listing of all publications and technical reports supported under this grant or contract.

(a)Papers published in peer-reviewed journals

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87. 83. John O'Hara and D. Grischkowsky, *Quasi-optic synthetic phased-array terahertz imaging*, J. Opt. Soc. Am. B., Vol. 21, 1178-1191 (2004).
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89. Jiangquan Zhang and D. Grischkowsky, *Adiabatic Compression of parallel-plate metal waveguides for sensitivity enhancement of waveguide THz time-domain spectroscopy (THz-TDS)*, Applied Physics Letters, Vol. 86, 061109-1-4, (2005).

(b) Papers published in non-peer-reviewed journals or in conference proceedings

1. Finite difference time domain studies of the local electric field enhancements near nanoshell structures, C. Oubre and P. Nordlander, *SPIE Proceedings* 5512(2004)28-37
2. Electronic structure and optical properties of metallic nanoshells, P. Nordlander and E. Prodan, *SPIE Proceedings* 5221(2003)151-163
3. Finite difference time domain studies of the optical properties of nanoshell structures, C. Oubre and P. Nordlander, *SPIE Proceedings* 5221(2003)133-143
4. Optical properties of metallic nanoshells, P. Nordlander and E. Prodan, *SPIE Proceedings* 4810(2002)91-98
5. C. E. Moran, Jennifer M. Steele, Allen Lee, Carla Aquirre, Corey Radloff, and N. J. Halas, "Soft Lithographic directed growth of wire grating arrays with optical resonances", *SPIE Proceedings* **4810**, 1-6 (2002).
6. Corey Radloff and N. J. Halas, "The decomposition of gold nanoshells in carbon tetrachloride", *SPIE Proceedings*, **4810**, 21-27 (2002).
7. J. B. Jackson and N. J. Halas, "Probing the Optical Near Field of a Nanolens", *SPIE Proceedings*, **4810**, 82-90 (2002).
8. Plasmon hybridization in metallic nanostructures, P. Nordlander, *SPIE Proceedings* 5512(2004)1-9

(c) Papers presented at meetings, but not published in conference proceedings:

<u>N. J. Halas:</u>
1. "Current Status and Future Trends in Near Field and Scanning Microscopies", NAS sponsored 2nd annual Japan-American Frontiers of Science Symposium (JAFOS), Tsukuba, Japan, October 1999.
2. "Metal Nanoshells", Graduate Research Seminar, ECE Department, Purdue University, October 1999.
3. "Nanotips and Nanoshells", Chemistry Colloquium, Rice University, October 1999.
4. "Metal Nanoshells: Properties and Interactions", Texas Section of the American Physical Society, University of Texas at Austin, October 1999.
5. "Oxidation Inhibition in Conducting Polymers-Gold Nanoshell Composites", Optical Probes 2000, Salt Lake City, UT, February 2000.
6. "Tailoring Novel Materials Properties with Composite Nanoparticles", Mechanical Engineering and Materials Science Department, Rice University, February 2000.

7. “Nanoengineering of Optical Properties: Applications and Commercialization of Metal Nanoshells”, Commercialization of Nanostructured Materials, Orlando, Florida, April 2000.
8. “Nanoshells and Applications”, Colloquium, Materials Science Department, Northwestern University, April 2000.
9. “Effects of Microgravity on Nanoparticle Growth”, NASA Microgravity Conference, Huntsville, AL, June 2000.
10. “Metal Nanoshell-polymer Composites: Changing the Properties of Functional Materials”, International Conference for Synthetic Metals, Bad Gastein, Austria, July, 2000.
11. “Metal Nanoshell-Polymer Composites”, ACS Regional Meeting, New Orleans, LA, December 2000.
12. “Nanoengineering Optical Properties with Metal Nanoshells: New Materials and Applications”, University of Arizona Optical Sciences Center, Tucson, AZ, March 2001.
13. “Metal Nanoshells: Materials Applications of Designer Nanoparticles”, Materials Science Seminar, University of Washington, Seattle, WA, March 2001.
14. “The Nanoengineering of Optical Properties: New Materials and Applications in Biotechnology”, Department of Electrical Engineering, University of Texas, Austin, TX, March 2001.
15. “Metal Nanoshells: Surface Chemistry and Dynamics”, Electrochemical Society Meeting, Washington, D.C., March 2001.
16. “Metal Nanoshells: Light Scattering Properties and their Applications Towards Sensor Protection”, DOD Sensor Protection Workshop, Army Research Laboratory, Adelphi, MD, March 2001.
17. “The Nanoengineering of Optical Properties using Metal Nanoshells”, Lawrence Livermore National Laboratory, Materials Research Institute Seminar, April 2001.
18. “Metal Nanoshells: Frequency Agile Optical Properties and Applications”, U. S. Air Force Research Laboratory, Wright Patterson AFB, Dayton, OH, May 2001.
19. “Metal Nanoshells: Shaping the Flow of Light One Nanoparticle at a Time”, Research Colloquium, University of California, Riverside, CA, May 2001.
20. “Metal Nanoshells: a Photonic Approach to Nanobiotechnology”, IBC Conference on Nanobiotechnology, San Diego, CA, July 2001.
21. “Nanotechnology and its Emerging Capabilities”, NCI sponsored workshop, NIST, Gaithersburg, MD, August 2001.
22. “Metal Nanoshells and Their Applications at the Nano-Bio Interface”, Red Herring Conference, Boston, MA, September 2001 (cancelled due to September 11 attacks).
23. “Metal Nanoshells: Shaping the Flow of Light One Nanoparticle at a Time”, ECE Colloquium, Cornell University, September 2001.
24. “Metal Nanoshells: Manipulating the Flow of Light at the Nanoscale”, Photonic Nanostructures Conference, Knowledge Foundation, San Diego, CA, October 2001.
25. “Properties of Metal Nanoshells”, 3M Corporation, St. Paul, MN, October 2001.
26. “Nanotechnology for the Real World”, Brother Lucian Blersch 2nd Annual Science Symposium, St. Edwards University, San Antonio, TX, Feb 2002.
27. “Metal Nanoshells: Constructing optics “from the dipole up”, Interdisciplinary Materials Science Graduate Program Colloquium, Vanderbilt University, Nashville, TN, April 2002.
28. “Nanoshells: Nanotechnology for the Real World”, Third Annual St. Olaf Honors Day Science Symposium- “Big Questions, Small Scale Solutions: New Frontiers in Nanoscience”, Northfield, Minnesota, May 2002.
29. “Tuning optical properties at the Nanoscale”, DAMOP May 28th-June 1st, 2002.
30. “Biophotonic applications of Metal Nanoshells”, Gordon Research Conference, “Lasers in Biology and Medicine”, July 2002.
31. “The Manipulation of Light one Nanoparticle at a time”, ETOPIM Conference, Snowbird, UT, July 2002.

32. “Nanoshells: bio-inspired architectures for multifunctional coatings”, ARL/AFRL Meeting on Multifunctional Coatings, Keystone, CO, August 2002.
33. “Nanoshells: a Photonic approach to Nanobiotechnology”, Texas Nano-Vivo Summit, Houston, TX, August 2002.
34. “Nanoshells: fabricating nanophotonics ‘from the dipole up’”, IEEE-Nanophotonics Colloquium, 2nd Annual IEEE Conference on Nanotechnology, August, 2002.
35. “Biomedical Applications of Gold Nanoshells”, 3rd Annual Bio-MEMS Conference, Columbus, OH, September 2002.
36. “Plasmonic Hybridization: Design Principles and Realization of Nanophotonic Architectures”, Physical Foundations of Quantum Electronics, Snowbird, UT, January 2003.
37. “Plasmonic Hybridization: Design Principles and Realization of Nanophotonic Architectures”, Research Seminar, University of Florida, January 2003
38. “Plasmonic Nanoparticles by Rational Design”, University of Georgia, January 2003
39. “ Plasmon hybridization: a Design Principle for the Realization of Practical Nanophotonic Architectures”, Georgia Institute of Technology, January 2003
40. “ Plasmon Hybridization: a rational design principle for nanophotonic architectures” Imperial College, UK, February 2003
41. Third International Conference on Biomimetic Materials Processing, BMMP-3; Nagoya, JP, January 26-30 2003
42. “Plasmonics: Rationally Designed Architectures for Optics at the Nanoscale”, Visit and Colloquium at Sandia National Laboratory, January 2003.
43. “Plasmonics: Rationally Designed Architectures for Optics at the Nanoscale”, Visit and Colloquium at Los Alamos National Laboratory, January 2003.
44. “Plasmonic Hybridization: Design Principles and Realization of Nanophotonic Architectures”, Australian Colloid and Interface Science 2003 meeting, Sydney, February 2003
45. “Nanoshells: using Tunable Plasmons for Nanobiotechnology”, Australian Nano-Bio meeting, Melbourne, February 2003
46. “Plasmonic Nanostructures and their Applications in Biosensing”, American Chemical Society meeting, March 2003
47. “Plasmon Hybridization: a design principle for the realization of practical nanophotonic architectures”, James Franck Institute Colloquium, University of Chicago, April 22nd, 2003
48. “Plasmon Nanostructures: Applications in Biotechnology”, Vanderbilt Student Invited Pharmacology Speaker, April 24, 2003.
49. “Plasmonics: Manipulation of electromagnetic fields at the nanoscale”, AFOSR Contractor’s meeting, San Diego, CA, May 18-22, 2003
50. “Nanoshells: an Ideal Nano-Bio Interface for Biomedical Applications”, Research Seminar, M. D. Anderson Cancer Center, June 2003
51. “Photothermal Effects Induced on and by Plasmonic Nanostructures”, Gordon Research Conference on Photothermal and Photoacoustic Phenomena, Colby-Sawyer College, NH, June 2003
52. “Nanoshells: an Ideal Nano-Bio Interface for Biomedical Applications”, AAMI Annual Meeting (American Association for Medical Instrumentation), Long Beach, CA, June 15, 2003
53. “Plasmonic Nanostructures by Rational Design”, Gordon Research Conference on Electronic Spectroscopy and Dynamics, July 2003
54. “Plasmonics: an Emerging Optical Nanotechnology”, First Annual SPRING meeting, Austin, TX, August 25-27.
55. “Optimizing Surface Enhanced Raman Scattering with Ag Nanoshells” J. B. Jackson and N. J. Halas, SPIE Conference, San Diego, CA, August 2003.

56. “Plasmonics: manipulating electromagnetic fields at the nanoscale”, Duke University, ECE Seminar, September 2003
57. “The Remarkable Optical Properties of Gold Nanoshells”, Gold 2003: Industrial Applications of Gold, International Conference, Vancouver, BC, September 2003
58. “Tunable Plasmonic Nanostructures: biosensing relevant properties and applications”, Biomedical Engineering Society (BMES) Annual Meeting, Nashville, TN, September 2003
59. “Plasmonics: Nanoscale Manipulation of the Plasmon Response”, Progress in Electromagnetic Research Symposium (PIERS2003), Honolulu, Hawaii, October 2003
60. . “Optics at the Nanoscale: Design Principles, Components and Applications”, Nanotechnology Symposium, Dartmouth College, November 2003
61. “Plasmonic nanostructures: Optical design at nanoscale dimensions”, International Symposium on Clusters and Nano-Assemblies (ISCANA), Virginia Commonwealth University, November 2003 (Plenary talk)
62. “Tunable Plasmonic Nanostructures: Fundamental Components for Nano-Optics”, Physics of Quantum Electronics 2004, Snowbird, UT, January 2004.
63. “Nanoshells: Tunable plasmonic nanoparticles with applications in biotechnology”, Inaugural Meeting of the Texas Academy of Science, Engineering and Medicine (TASEM), San Antonio, TX, January 2004.
64. “Nanophotonics in Silica/Gold Nanoshells”, Contemporary Photonics Technology 2004, Tokyo, Japan, January 2004
65. “Nanoshells: Optical Design at Nanoscale Dimenions”, Research Seminar, Nanophotonics Laboratory, Professor Satoshi Kawata, University of Tokyo, Japan, January 2004.
66. Plenary Talk, 3rd Annual Rice Alliance Nanotechnology Innovation Forum, Rice University, January 2004 (declined due to conflict).
67. SPIE Biophotonics Meeting, “Plasmonic Applications in Biomedicine”, (talk declined, Rebekah Drezek suggested as substitute speaker to present nanoshell-related work).
68. “Plasmon Hybridization: a Design Principle for the Realization of Practical Nanophotonic Architectures”, Physics Department Colloquium, University of Colorado, January 2004
69. “Nanoshells: design rules and chemical fabrication of tunable nanophotonic components”, Chemistry Department and IGERT Seminar, University of Oregon, February 2004
70. IEEE Nanoscale Devices and Systems Integration, Orlando, FL, February 2004 (declined due to conflict)
71. “Nanoshells: Fundamental Plasmonic nanocomponents with Real-world Applications” Research Seminar, Zyvex Corp. (televised), Dallas, TX, February 2004.
72. “Nanoshells: Optical Design at Nanoscale Dimensions”, Physics Colloquium at Trinity University, San Antonio, TX, February 2004.
73. “Nanoshells: fundamental topologies and design principles for nano-optics”, Physics Colloquium, Rice University, Houston, TX, March 2004.
74. Plenary talk, 113rd TMS Annual Meeting (Minerals, Metals and Materials), Charlotte, NC, March 2004 (declined due to travel conflict)
75. PIERS2004 Annual Meeting, Pisa, Italy, March 2004 (talk presented by Glenn Goodrich).
76. “Nanoshells: New tools for manipulating light at the nanoscale”, Nanotech 2004 Conference, Tokyo, Japan, March 2004 (Plenary talk)
77. “Nanoshells: New tools for manipulating light at the nanoscale”, NAE Regional Meeting, Rice University, March 2004.
78. “Nanoshells as Multimodality nanoscale sensors”, 2004 ACS National Meeting, Anaheim, CA March, 2004.
79. “Plasmonic Nanoparticles by Rational Design”, 2004 ACS National Meeting, Anaheim, CA, March 2004.
80. “Tailoring the near field for enhanced spectroscopies below the diffraction limit”, 2004 ACS National Meeting, Anaheim, CA, March 2004.

81. Workshop on Nano-scale Materials: From Science to Technology, Institute of Physics, Bhubaneswar, India during April 5-8, 2004 (declined).
82. “Tailoring nanostructures for enhancing spectroscopies below the diffraction limit”, MRS Spring Meeting, San Francisco, CA April 2004.
83. “Rational Design of Plasmonic Nanoarchitectures”, MRS Spring Meeting, San Francisco, CA April 2004
84. “Symmetry Breaking in Synthesis and Post-synthetic Processing of Nanoshells”, MRS Spring Meeting, San Francisco, CA April 2004
85. ACI Nanobiotechnology Conference, San Francisco, CA, April 2004 (declined due to conflict).
86. “Nanoshells: using Nanotechnology to harvest light for Biomedicine”, Benson Lecturer, Physics Department, Miami University, April 2004 (Public Lecture).
87. “Plasmonic Nanostructures by Rational Design”, Benson Lecturer, Physics Department, Miami University, April 2004 (Physics Colloquium).
88. “ Nanoshells: Tunable Plasmonic Nanostructures by Rational Design”, Women in Science and Engineering Distinguished Lecture and Physics Colloquium, Kansas State University, Manhattan, Kansas, April 2004.
89. “Nanoshells: applications of Plasmonic Nanostructures in Biomedicine” NIH/NIAID Workshop, Gaithersburg, MD, June 2004.
90. “Nanoshells: Nanoscale Manipulation of the Plasmon Response”, EMRS (European Materials Research Society), Strasbourg, France, May 2004.
91. “Nanoshells: from plasmon physics to cancer therapy”, Research Seminar, Chalmers University, Goteborg, Sweden, June 2004.
92. Association of Academic Health Centers, Council on Research and Science, Washington, DC (declined due to conflict).
93. Frontiers in Laser Physics, Trieste, IT, July 14-16, 2004.
94. “Photonics from the Bottom Up: Design Tools for Manipulating Light at the Nano Scale”, Chemistry and Physics of Nanostructure Fabrication, Gordon Research Conference, Tilton, NH, July 18-23, 2004.
95. Invited talk, Nanophotonics Conference, Osaka, Japan, July 2004.
96. Invited talk, SPIE Annual Meeting, Denver, CO, August 2004.

<u>P. Nordlander</u>

1. Plasmon hybridization in complex metallic nanostructure, SPIE conference on Optical Science and Technology, 8/2/05, San Diego
2. Nanoplasmonics and surface enhanced spectroscopies, 4:th International workshop on surfaces, Beijing, PRC, 5/31/05
3. Plasmons and surface enhanced spectroscopies, Kyushu University, Japan, 5/17/05
4. Optical properties of metallic nanostructures, ICU Tokyo, Japan, 5/16/05
5. Plasmons in metallodielectric nanostructures, NIST Gaithersburg, 4/29/05
6. Plasmon hybridization in nanostructures, 34:th Symposium on the Physics of Quantum Electronics, Snowbird, 1/7/04
7. Nanoparticle enhanced spectroscopies, DAMOP, Tucson, 5/26/04
8. Plasmon hybridization in metallic nanostructures, SPIE conference on Optical Science and Technology, 8/2/2004, Denver
9. Plasmons in metallodielectric nanostructures, Texas A&M, 9/15/04
10. Plasmons in metallic nanostructures, LLNL, Livermore, 10/14/04
11. Electronic structure and optical properties of metallic nanoshells, Progress in Electromagnetics Research Symposium (PIERS2003) Waikiki 10/14/03
12. Electronic structure and optical properties of metallic nanoshells, SPIE conference on Optical

Science and Technology, San Diego 8/5/03
13. Electronic structure and optical properties of metallic nanoshells, UCLA 4/28/03
14. Electronic structure and optical properties of metallic nanoshells, ACS meeting New Orleans 3/19/03
15. Electronic structure and optical properties of metallic nanoshells, APS meeting Austin March 3/5/03
16. Plasmons and optical properties of metallic nanoshells, Army Research Laboratory, Adelphi MD 9/11/02
17. Electronic structure and optical properties of metallic nanoshells, Giambiagi 2002, Buenos Aires, 7/24/02
18. Current topics in nano-science and nano-technology, Fuji Research Laboratories, Tokyo, 5/17/01

(d) Manuscripts submitted, but not published (none)

(e) Technical reports submitted to ARO (none)

(7) List of all participating scientific personnel showing any advanced degrees earned by them while employed on the project:

Rice University

Prof. Naomi J. Halas
Prof. Peter J. Nordlander
Prof. Alexander J. Rimberg
Prof. Jennifer West

Dr. Sandra Bishnoi
Dr. Shiqing Man

Carla Aguirre – M.S. - 2004
Bruce Brinson
Taylor M. Cavanah
Joseph Cole
Nathaniel Grady
Leon R. Hirsch
G. D. Hale – Ph.D. – 2000
Joseph B. Jackson – M.S. - 2000, PhD – 2004
Surbhi Lal
Nikolay A. Mirin
Cristin E. Moran – Ph.D. May 2004
Colleen Nehl – M.S. - 2005
Christopher D. Oubre – M.S. - 2003, PhD - 2005
Emil V. Prodan – Ph.D. - 2003
Corey Radloff – Ph.D. - May 2004
Scott R. Sershen
David Sicilia
Jennifer M. Steele – M.S. - 1999, Ph.D. - 2004
SriPriya Sundararajan
Felicia Tam – M.S. - 2005
Sarah L. Westcott – Ph.D. – 2001

John A. Wolfgang – M.S. - 2000, Ph.D. - 2001

Oklahoma State University
Prof. R. A. Cheville
Prof. D. Grischkowsky

M. T. Reiten – Ph.D. expected 2002
Stacee A. Harmon – M. S. - 2002
Shyla Krishnamurthy – M. S. - 2002
John O'Hara – Ph.D. - 2003
R. Mendis – Ph.D. - 2001

(8) Report of Inventions (by title only)

Temperature-Sensitive Polymer/Nanoshell Composites for Photothermally Modulated Drug Delivery 6,428,811 and 6,645,517with Jennifer L. West, Scott R. Sershen, Steven J. Oldenburg, and Richard D. Averitt.

Partial Coverage metal nanoshells and method of making same, 6,660,381 with Robert K. Bradley

Metal Nanoshells for biosensing applications, 6,699,724 with Jennifer West, Steve Oldenburg and Richard D. Averitt

Nanoparticle-based all-optical sensors, 6,778,316 with Surbhi Lal, Peter Nordlander, Joseph Jackson, and Cristin Moran

(9) Bibliography
(10) Appendixes